

PROCESS DEVELOPMENT STUDIES OF
TWO-STAGE LIQUEFACTION AT WILSONVILLE

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INTRODUCTION

The Advanced Coal Liquefaction R&D Facility at Wilsonville, Alabama, has been in operation for over 12 years. It is the largest direct coal liquefaction pilot plant still in operation in the United States. Process investigations have evolved from the original study of the Solvent Refined Coal Process for making a clean solid fuel to the recent investigation of two-stage liquefaction processes for making clean distillate fuels. This paper presents results from the current study of various processing schemes designed to reduce the cost of fuels produced by two-stage liquefaction plants.

Most important among these configurations is direct coupling of the thermal and catalytic reactors. Such close-coupled operation should lower the cost of two-stage fuels by increasing overall thermal efficiency and improving product quality. Results from Wilsonville runs are characterized by discussion of representative product yield and product quality data. Also, a comparison of performance and stability of Shell 324 and Amocat 1C catalysts is presented.

Other pertinent tests in the close-coupled mode are discussed with particular emphasis on the effect of higher system space velocity and on the impact of solids recycle.

PROCESS CONFIGURATION COMPARISON

The integrated TSL (ITSL) mode and the close-coupled (CC-ITSL) mode are shown in Figures 1 and 2, respectively. In both modes, the reaction stages are integrated by the recycle of hydrotreated resid with the goal of producing "all-distillate" yield slates. The CC-ITSL configuration differs from ITSL in that for the CC-ITSL mode, products from the thermal reactor are fed directly to the catalytic reactor without pressure letdown or significant cooling. Accordingly, vacuum-flashed product from CC-ITSL is deashed downstream of the catalytic reactor rather than upstream as in the ITSL mode. Close-coupled operation thus eliminates thermal inefficiency of the cool down/reheat cycle associated with deashing between reaction stages.

PROCESS PERFORMANCE

A wide range of thermal and catalytic stage operating conditions were investigated in the CC-ITSL mode to determine their effects on product yield structure and quality. For comparison purposes, Table 1 lists five sets of CC-ITSL conditions and yields relative to one typical set of ITSL conditions and yields. Illinois No. 6 coal from Burning Star Mine was used in all the runs.

An initial baseline run (250B) was conducted using aged Shell 324 catalyst to quantify the impact of coupling the reactors. No interstage vapor separation was utilized in Run 250B, although a high pressure separator was installed and operated during the remainder of the run. The major effect of close-coupled operation was an increase in hydrogen consumption with a corresponding improvement in distillate product quality (Table 2). Hydrogen and sulfur values were significantly better for CC-ITSL (250B) when compared to ITSL (244). This improvement resulted from the fact that all the TSL product was derived from the

catalytic stage in Run 250B, whereas in Run 244 approximately 45 wt % was produced by the catalytic hydrotreater (Table 3). Higher quality products are made by the catalytic stage.

Subsequent operation in CC-ITSL was devoted to study of performance of a new catalyst, Amocat 1C. As noted previously, interstage vapor separation was employed throughout this period. A comparison of "all-distillate" operation at the same coal feed rate for Shell 324 and Amocat 1C revealed that distillate and hydrocarbon gas yields were similar (Run 250B versus Run 250C). An alternate deashing solvent was used in Run 250C in order to maintain stable performance while processing the highly soluble feed generated by the fresh Amocat catalyst. This resulted in greater rejection of resid to ash concentrate with concomitantly less TSL resid produced. A lower hydrogen consumption was observed for Run 250C. This phenomenon could be attributed to the effect of interstage separation (i.e., not hydrotreating all distillate products) and/or to relatively lower hydrogenation activity of the Amocat catalyst.

Higher system space velocities were explored by increasing coal feed rates. The goal was to demonstrate the production of high quality (CC-ITSL) distillates at reduced cost. Yield data are reported for Run 250 Periods C, D, and E in Table 1. Product quality data and unit contributions to distillate production for Period D are given in Tables 2 and 3, respectively. The data clearly indicate that "all-distillate" yield slates were produced at increased throughputs by compensatory increases in reactor temperatures. Product quality did not change significantly at the higher rates. It should also be noted that products from higher space velocity CC-ITSL were substantially better than those from lower space velocity ITSL.

Near the conclusion of Run 250, a test of solids (unconverted coal and ash-cresol insolubles) recycle was performed (Figure 3). The objective was to decrease size of the critical solvent deashing unit by deashing a higher solids content vacuum bottoms stream. Deashed resid was recycled as a component of the liquefaction solvent. Lower organic rejection to the ash concentrate was demonstrated with the concentrated feed. Based on this result, the concept of solids recycle may be investigated in a future close-coupled run using catalyst in both reactors.

Batch deactivation trends for resid conversion in the catalytic reactor were developed for the ITSL (deashed bituminous)-Shell 324 and CC-ITSL (close-coupled bituminous)-Amocat 1C modes, using a first order resid conversion model (1). The trends are plotted in Figure 4 together with batch deactivation data from Wilsonville Run 247 ("simulated" close-coupled). The trends showed initial periods of rapid deactivation, followed by slower deactivation rates. Higher resid conversion rate constants were observed for close-coupled operation using the Amocat 1C catalyst. The close-coupled resid/Amocat combination was more reactive than the other feed/catalyst (Shell 324) combinations. Figure 5 further illustrates this point in an Arrhenius plot.

RELATIVE ECONOMICS

Results from an economic screening study performed by Lummus Crest Inc. indicated a reduction in the required product selling price for CC-ITSL products compared to ITSL products (Table 4). The study was based on a conceptual commercial 10,000 TPD plant using Illinois No. 6 coal. The relatively lower price was due to higher distillate production and improved distillate quality for the close-coupled case (2).

SUMMARY

- The major effect of close-coupled operation was an increase in hydrogen consumption with a corresponding improvement in distillate product quality.
- At the same coal feed rate, C₄+ distillate and hydrocarbon gas yields were similar for close-coupled operation with Shell 324 and Amocat 1C catalysts. A lower hydrogen consumption was observed in the Amocat operation. This phenomenon could be attributed to the effect of interstage separation and/or to the relatively lower hydrogenation activity of the Amocat catalyst.
- "All-distillate" yield slates were produced at higher system space velocities by compensatory increases in reactor temperatures.
- Product quality was not significantly affected at higher system space velocities.
- Operating with solids recycle in the close-coupled mode, lower organic rejection to ash concentrate was demonstrated at a relative reduction in feed rate to the critical solvent deashing unit.
- A comparison of resid conversion rate constants indicated that the close-coupled resid/Amocat 1C combination was more reactive than other feed/catalyst (Shell 324) combinations.
- Results from an economic screening study for a conceptual commercial 10,000 TPD plant indicated a reduction in the required product selling price for CC-ITSL plant products compared to ITSL plant products.

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REFERENCES

1. Rao, A. K., Gadiyar, H. J., Pate, F. L., "Catalytic Hydrogenation of SRC-I Product at the Wilsonville Pilot Plant", Proceedings of the Seventh Annual EPRI Contractors' Conference on Coal Liquefaction, May 1982.
2. Correspondence from M. Peluso to W. Weber, "RP832-11, Final Technical and Economic Data", January 23, 1986.

Table 1

PROCESS CONDITIONS AND YIELDS

Configuration Run No.	ITSL 244	CC-ITSL(1) 250B	CC-ITSL(2) 250C	CC-ITSL(2) 250D	CC-ITSL(2) 250E	CC-ITSL(2) 250F
Catalyst Type	Shell 324	Shell 324	Amocat 1C	Amocat 1C	Amocat 1C	Amocat 1C
Thermal Stage						
Reactor temperature (°F)	810	810	810	825	830	835
Coal space velocity [lb/hr-ft ³ (>700°F)]	28	25	25	37	45	37
Inlet H ₂ partial pressure (psi)	2040	2800	2380	2380	2380	2380
Solvent-to-coal ratio	1.8	1.8	1.8	1.8	1.8	1.9(5)
Catalytic Stage						
Reactor temperature (°F)	730	730	700	740	750	760
Space velocity (lb feed/hr-lb cat)	1.0	1.0	1.3	2.1	2.6	2.2
Catalyst age (lb resid/lb cat)	1070	1450(3)	350(3)	750(3)	1100(3)	550(3)
TSL Yield(4)(% MAF Coal)						
C ₁ -C ₃ Gas	6	6	5	7	6	8
C ₄ + Distillate	57	61	61	64	58	62
Resid	7	6	2	-2	7	7
Ash Concentrate (ash-free)	20	18	22	19	19	15
Hydrogen Consumption	-5.4	-6.5	-5.6	-6.1	-5.7	-6.5

(1) Without interstage vapor separation.

(2) With interstage vapor separation.

(3) Catalyst age is (lb resid + ash + unconverted coal)/lb catalyst.

(4) Elementally balanced yield structure.

(5) Recycle solvent contains unconverted coal and ash (cresol insolubles-CI).

Table 2

PROPERTIES OF DISTILLATE PRODUCTS

Distillation Cut	Wt % of Crude	Elemental (Wt %)				S	O (diff)
		C	H	N			
ITSL (Run 244) (1)							
Naphtha (TBP-350°F)	18.6	85.21	12.86	845 ppm		0.36	1.50
Distillate (350-650°F)	46.1	86.34	10.73	0.23		0.22	2.48
Gas Oil (650°F+)	35.3	89.07	9.69	0.31		0.16	0.76
CC-ITSL (Run 2508) (2)							
Naphtha (TBP-350°F)	19.0	85.31	14.51	0.17		0.01	-
Distillate (350-650°F)	46.8	87.43	12.12	0.21		0.06	0.18
Gas Oil (650°F+)	34.2	88.41	10.88	0.57		0.04	0.10
CC-ITSL (Run 2500) (3)							
Naphtha (TBP-350°F)	18.6	85.35	14.25	0.22		0.18	-
Distillate (350-650°F)	45.0	87.45	11.73	0.32		0.09	0.41
Gas Oil (650°F+)	36.4	88.77	10.27	0.43		0.07	0.46

(1) Samples obtained with catalytic stage temperature of 730°F, space velocity of 1.0 lb feed/hr-lb catalyst, and catalyst age of 1400 lb resid/lb catalyst (Shell 324).

(2) Samples obtained with catalytic stage temperature of 730°F, space velocity of 1.0 lb feed/hr-lb catalyst, and catalyst age of 1450 lb (resid + ash + unconverted coal)/lb catalyst (Shell 324).

(3) Samples obtained with catalytic stage temperature of 740°F, space velocity of 2.1 lb feed/hr-lb catalyst, and catalyst age of 750 lb (resid + ash + unconverted coal)/lb catalyst (Amocat 1C).

Table 3

UNIT CONTRIBUTIONS TO DISTILLATE PRODUCTION

Configuration Run No. Catalyst Type	ITSL 244 Shell 324	CC-ITSL(1) 250B Shell 324	CC-ITSL(2) 250D Amocat 1C
<u>Unit Contributions(3)</u>			
Thermal Stage	55	0	25
Catalytic Stage	45	100	75

(1) Without interstage vapor separation.

(2) With interstage vapor separation.

(3) Wt % of total distillate production.

Table 4

ECONOMIC SCREENING STUDY(1)
(10,000 TPD PLANT)

	ITSL (BASE)	CC-ITSL
Relative Capital Cost	1.00	1.04
Relative Operating Cost	1.00	1.01
Relative Production Rate(2)	1.00	1.10
Relative Required Product Selling Price(3)	1.00	0.93
Thermal Efficiency (%)	68.1	69.6

(1) Performed by Lummus Crest, Inc. under EPRI RP832-11.

(2) Based on barrels of crude oil equivalent with consideration for quality of distillate products.

(3) First year.

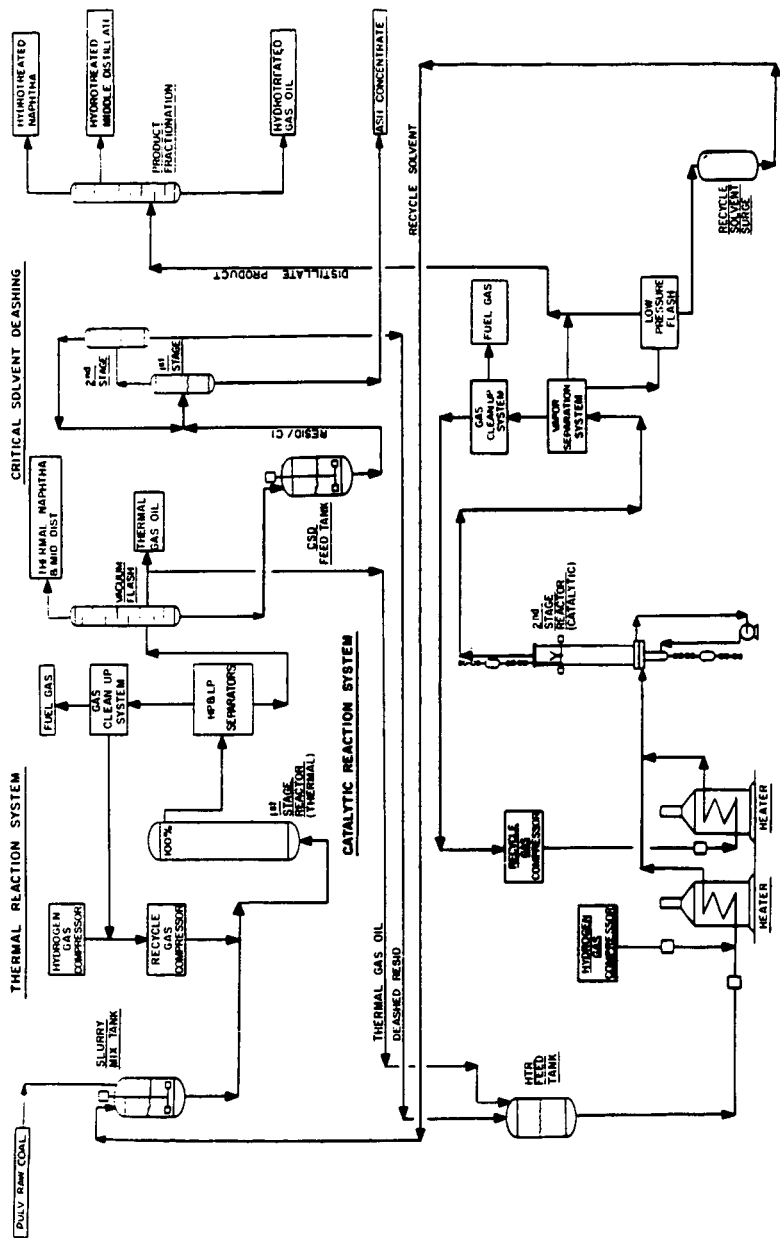


FIGURE 1: INTEGRATED TWO STAGE LIQUEFACTION SYSTEM
INTER-STAGE DEASHING

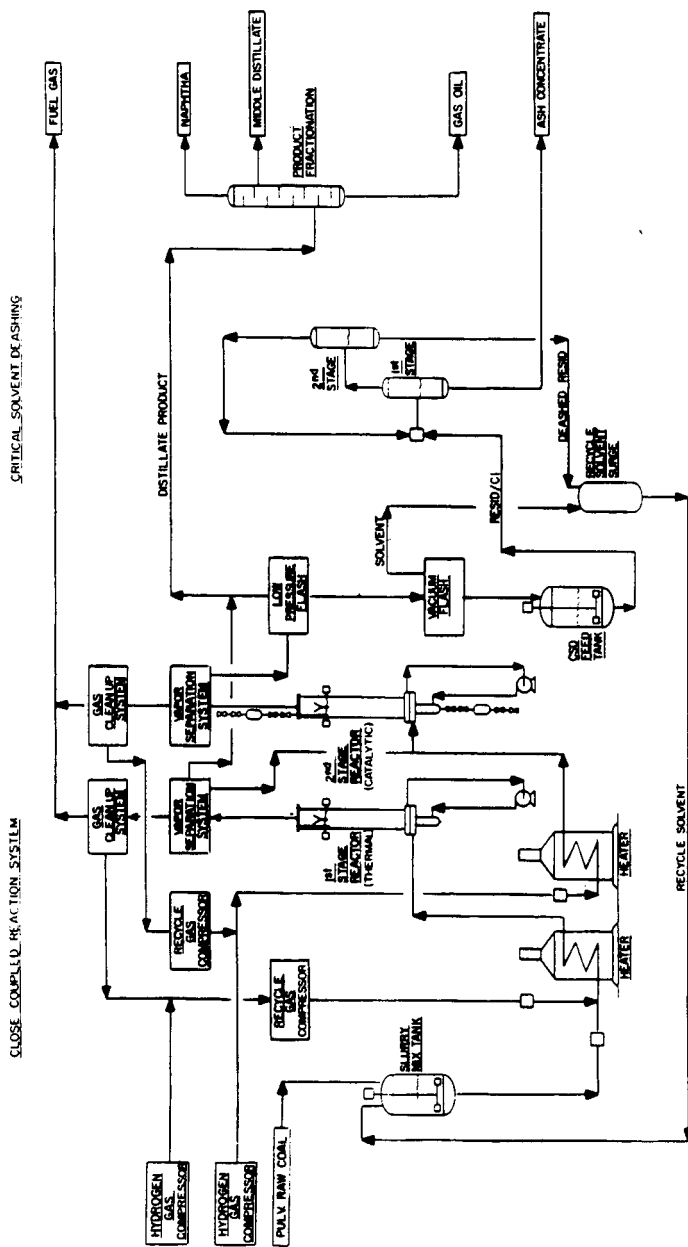


FIGURE 2: CLOSE COUPLED INTEGRATED TWO STAGE LIQUEFACTION SYSTEM
THERMAL - CATALYTIC WITH INTER-STAGE VAPOR SEPARATION

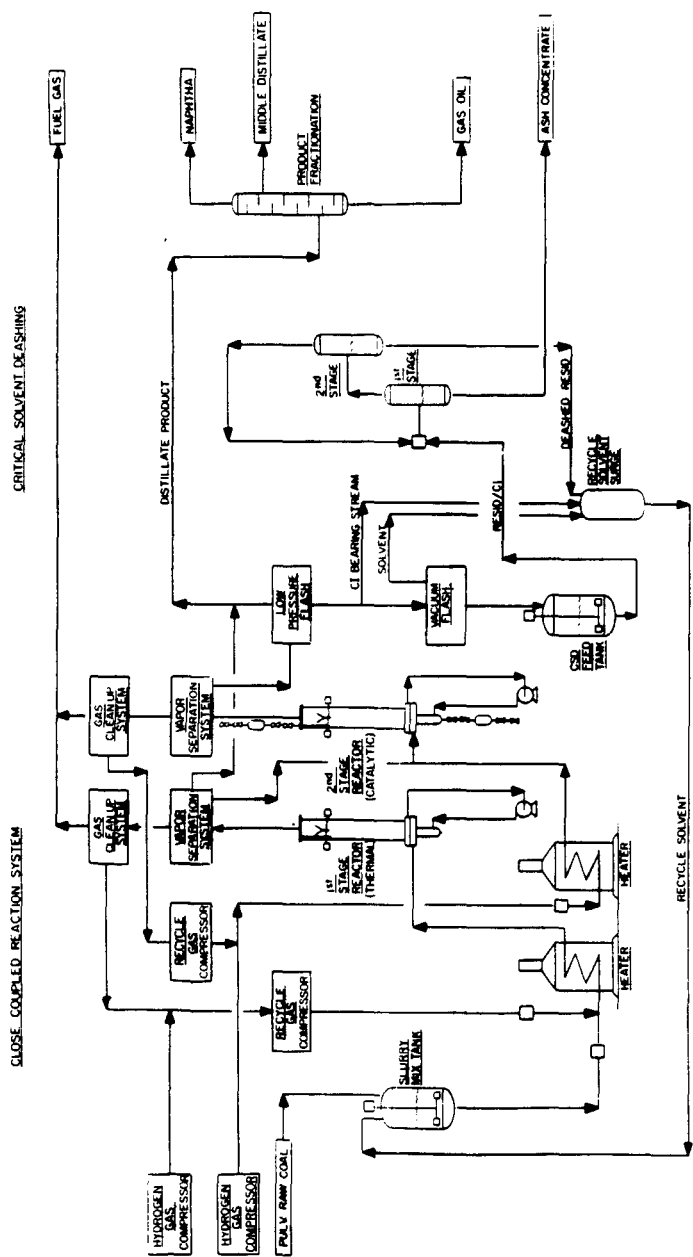


FIGURE 3. CLOSE COUPLED INTEGRATED TWO STAGE LIQUEFACTION SYSTEM
THERMAL - CATALYTIC WITH CI RECYCLE

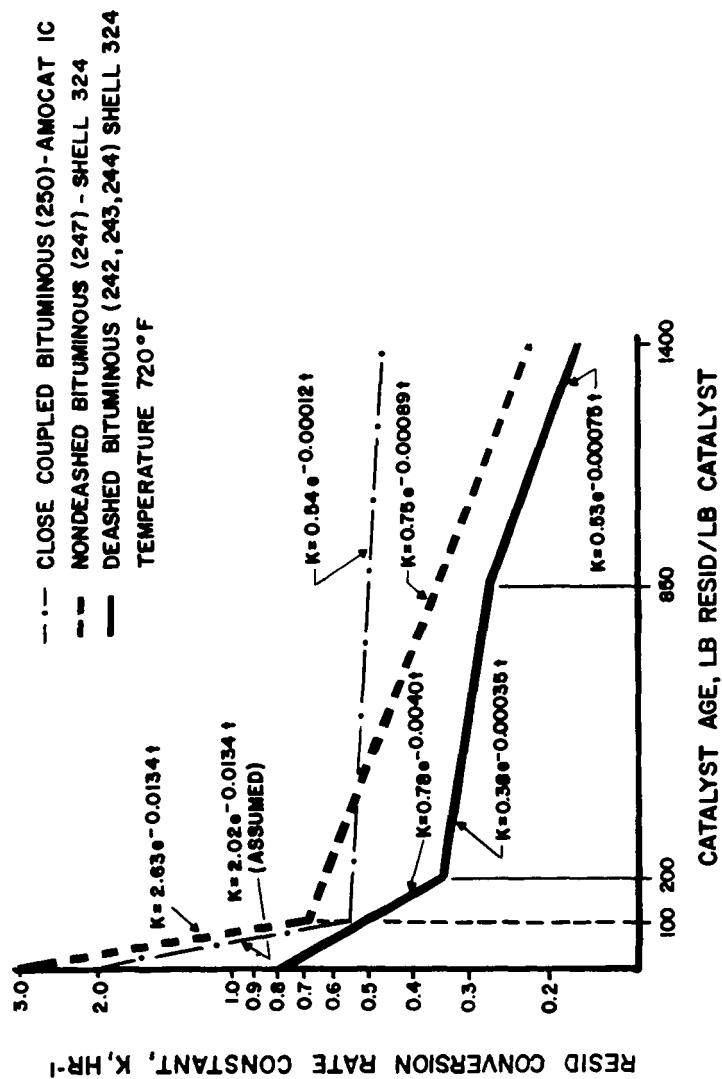


FIGURE 4. CATALYST DEACTIVATION TRENDS

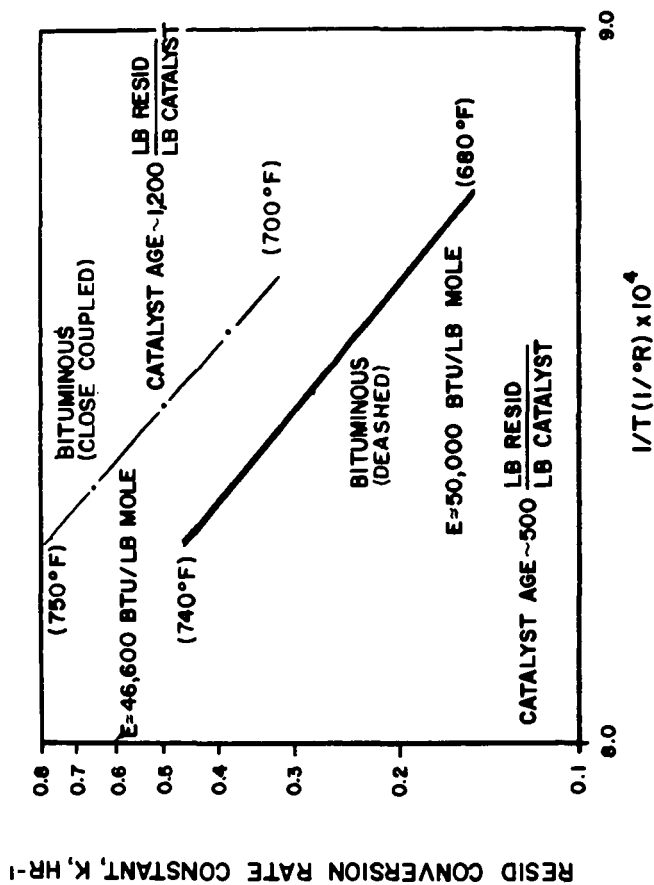


FIGURE 5. COMPARISON OF RESID CONVERSION RATE CONSTANTS